

# SINGLE-SHOT MULTI-MeV ULTRAFAST ELECTRON DIFFRACTION ON VELA AT DARESBUURY LABORATORY

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## Abstract

Accelerator-based Ultrafast Electron Diffraction (UED) is a technique for studying both static structure and sub-100 fs dynamic structural changes on the atomic scale. In this paper we present the first electron diffraction results, obtained in 2014, on the Versatile Electron Linear Accelerator (VELA) at Daresbury Laboratory. Diffraction patterns were observed with much less than 1 pC transported to the detection screen. Single-shot and multi-shot accumulated diffraction data are presented from single crystal and polycrystalline samples. Apertures of different sizes placed directly in front of the sample offer some control over the contamination due to dark current, but also reduce the charge contributing to the diffraction pattern. We discuss future developments for electron diffraction on VELA, including further beam optimisation, the measurement of bunch length with a newly installed Transverse Deflecting Cavity (TDC), and the plans for pump-probe studies.

## INTRODUCTION

Structural science, particularly structural biology, is a major driver in the development of new light sources. However, many important materials such as membrane proteins and protein complexes cannot be crystallised into sufficiently large crystals for use in conventional X-ray diffraction arrangements and moreover, are often damaged by the beam before the data collection can be completed. A new approach is to use X-ray Free-Electron Lasers (FELs) to produce ultra-high intensity sub-100 fs X-ray pulses to allow single shot diffraction data to be recorded before the sample deteriorates, that is, the data collection is faster than the radiation damage. This approach is being pursued at SACLA [1] and LCLS [2]; for example LCLS have recently successfully carried out X-ray diffraction imaging of a giant Mimivirus [3].

Structural evolution on a time-scale comparable to that of the making and breaking of chemical bonds can also be studied with these high intensity X-ray FELs. Recently, ultrafast resonant soft X-ray diffraction experiments at LCLS allowed the optically stimulated changes of charge-density wave correlations in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$  to be followed [4].

An attractive alternative approach to using X-ray FELs to study structural changes is ultrafast electron diffraction. Achieving the femtosecond time resolution needed to observe dynamic changes in molecules requires the use of multi-MeV electrons to reduce space charge effects that lengthen the bunch.

Electrons offer several advantages over X-rays for diffraction studies:

(1) Electrons have a Coulombic interaction with both the electrons and the nuclei of the target atom whilst X-rays only scatter from the electrons. Electrons thus have a scattering cross-section which is 4-6 orders of magnitude larger than that of X-rays.

(2) Electrons have a lower ratio of inelastic to elastic scattering cross-sections than do X-rays.

(3) Electrons deposit a lower energy into the target during inelastic collisions than do X-rays [5-7].

The net result is that electrons deposit three orders of magnitude less energy per useful scattering event than do X-rays, thereby reducing the damage done to the sample. While it is frequently argued that a 10 fs X-ray pulse will “beat” Coulomb explosion of the molecule, this may not be the case as the electron density from which X-rays are scattered is being disrupted on much faster (attosecond) timescales. In contrast, electrons scatter from the nuclei and so are much less susceptible to damage mechanisms which remove electron density. Ultrafast electron diffraction also has notable cost advantages over FELs for diffraction experiments since much smaller scale accelerators, such as VELA [8], can be used.

## UED ON VELA

A description of the electron diffraction system on VELA has been presented elsewhere [9] where it was shown that, although sub-100 fs 1 pC bunches can be generated at the gun, the bunch length increases as the beam propagates along the accelerator. A schematic of the accelerator is shown in Fig. 1. For the electron diffraction experiments described in this article, VELA was operated at 4 MeV/c. Seven quadrupole magnets, located between the gun and the sample chamber, are used to shape the electron beam. The beam can be imaged on YAG screens at several places along the accelerator, including the sample position.

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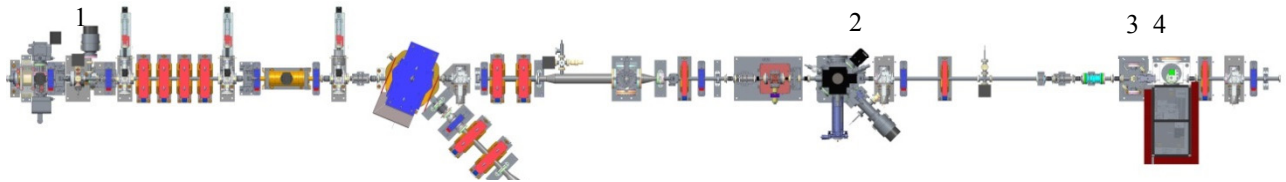


Figure 1: VELA schematic layout. {1} Photocathode gun, {2} UED chamber, {3} Insertable Faraday cup, {4} Detector chamber and CCD

The shape of the electron beam immediately in front of the sample can be controlled remotely by inserting circular apertures, with diameters ranging from 0.5 to 5 mm, into the beam. The VELA gun produces dark current at the required field gradients and the apertures can be used to reduce this contribution to a very low level. The accelerator delivered typically 1 pC of charge from the gun but the apertures removed the majority of this leaving typically ~40 fC at the sample.

From the sample, the beam travels 3.4m to a Lanex detection screen. The diffraction pattern was imaged, via a 45° mirror, with an iXon3 image-intensifying camera fitted with a Samyang 85mm f/1.4 lens and capable of single photon detection. A central hole in both the screen and the mirror removes the high intensity undiffracted beam. A second screen is available for investigating the undiffracted beam. A remotely-controlled Faraday cup can be inserted to measure the charge at the screen.

## RESULTS

A range of samples, including polycrystalline platinum, gold, aluminium, graphite particles and single crystal gold (Au (100)), was used to demonstrate electron diffraction on VELA. All the samples were supported on Ni or Au TEM grids and mounted in a carousel which allowed them to be interchanged. The carousel is capable of holding two samples of up to 6 mm in diameter and six samples on standard 3.05 mm diameter TEM grids (Fig. 2).

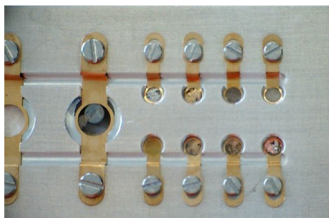


Figure 2: Sample carousel.

The first results were obtained using a polycrystalline platinum sample which was estimated to be 20 nm thick. The diffraction pattern, obtained as a sum of 1000 shots and after subtraction of the dark current background, is shown in Fig. 3. The background for subtraction was obtained simply by closing the photoinjector laser shutter whilst maintaining the RF field.

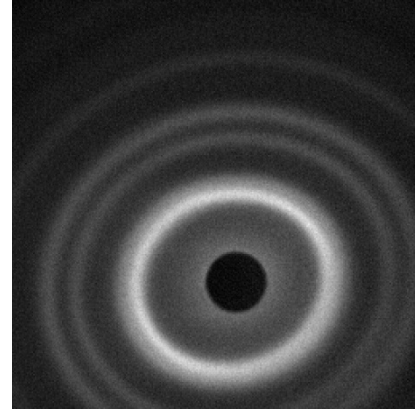


Figure 3: Diffraction pattern from polycrystalline platinum, obtained as the sum of 1000 shots at < 1 pC.

The intensity of the diffracted beam was obtained from the azimuthal integration of the diffraction image shown in Fig. 3. A fitted baseline was then subtracted from the integrated image, resulting in Fig. 4. For these early results, the FWHM of the combined (111) and (200) diffraction peak is  $\Delta s \approx 0.77 \text{ \AA}^{-1}$ .

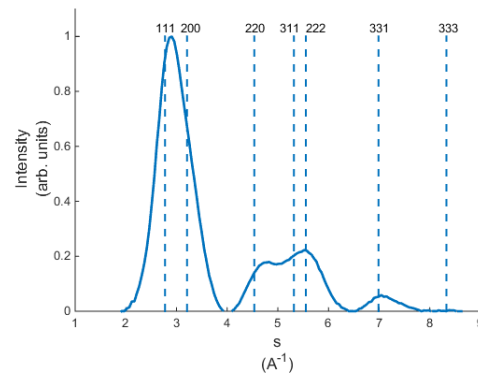


Figure 4: Measured polycrystalline diffraction pattern (solid line) with expected positions of diffraction peaks overlaid (dashed lines) for platinum.

Results are expressed as a function of the magnitude of momentum transfer between the incident electron in the beam and the scattered electron;  $s = \frac{4\pi}{\lambda} \sin\left(\frac{\theta}{2}\right)$ , where  $\theta$  is the scattering angle and  $\lambda$  is the de Broglie wavelength of the electrons.

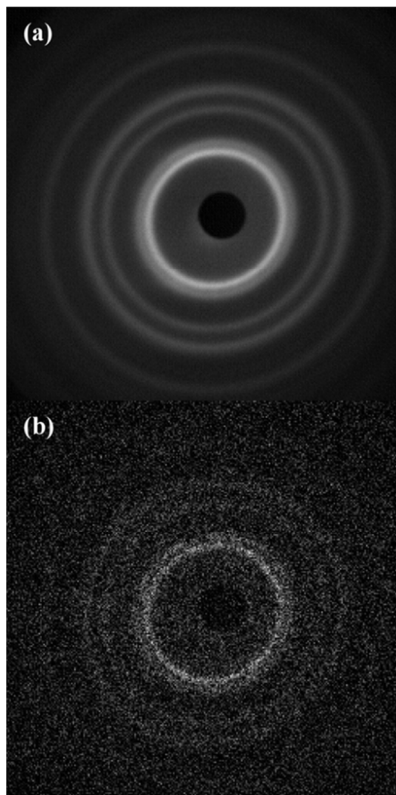


Figure 5: Diffraction patterns from polycrystalline gold (a) 1000 accumulated shots at 40 fC (b) single-shot at 40 fC.

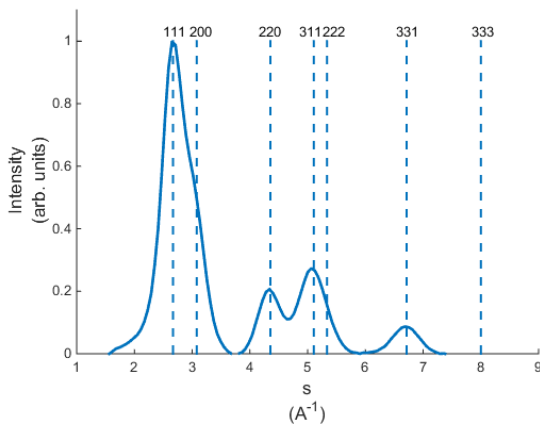


Figure 6: Azimuthal integration of the polycrystalline gold pattern in Fig. 5a (solid line) with expected positions of diffraction peaks overlaid (dashed lines).

Figure 5 shows diffraction patterns from polycrystalline gold recorded after refining the machine set up. A 1 mm diameter aperture in front of the sample reduced the charge to  $\sim 40$  fC. Even at this low charge, a single-shot pattern can be successfully recorded. The (111) and (200) diffraction peaks are still merged (Fig. 6), but are more easily distinguishable than those in Fig. 4. Figure 6 also shows that the observed pattern correlates with the expected peak positions. The FWHM of the combined (111)/(200) diffraction peak is  $\Delta s \approx 0.63 \text{ \AA}^{-1}$ .

Single crystal diffraction was also demonstrated from a (100) orientated gold crystal, and a single-shot diffraction pattern is shown in Fig. 7.

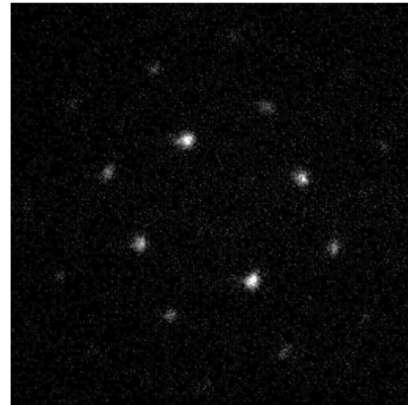


Figure 7: Diffraction pattern from single crystal gold obtained with a single 40 fC electron pulse. The central undiffracted spot is absent due to the hole in the screen.

## FUTURE DEVELOPMENTS

A successful UED facility needs to minimise the bunch length to give the best temporal resolution. A Transverse Deflecting Cavity (TDC) was recently installed and commissioned to allow the bunch length to be measured accurately [10] and will be used to optimise the low-charge operation of the machine. Reducing the space-charge induced increase in bunch length will be achieved by reconfiguring VELA and moving the sample chamber closer to the gun. We plan to incorporate the diffraction processes into the full start-to-end simulations of the machine to determine optimised operating parameters.

The long-term goal is to develop a pump-probe scheme. A light pulse from an optical laser would be used to induce a change in the sample, e.g. excitation into an excited electronic state. UED will then be used to record the time evolution of the subsequent changes. The ultimate aim is to study reactions in non-equilibrium states of matter as a function of time.

## CONCLUSION

Single-shot diffraction patterns from polycrystalline and single crystal samples have been recorded on VELA with electron pulses having charges of a few tens of fC. A programme of machine optimisation and characterisation will allow dynamic structural studies to be made on the sub-100 fs timescale.

## ACKNOWLEDGMENTS

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